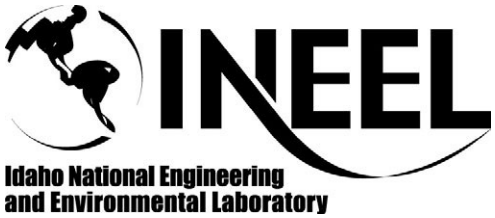

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PREPRINT

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Performance of Thorium-Based Mixed Oxide Fuels for the Consumption of Plutonium in Current and Advanced Reactors

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Abstract – *A renewed interest in thorium-based fuels has arisen lately based on the need for proliferation resistance, longer fuel cycles, higher burnup and improved wasteform characteristics. Recent studies have been directed toward homogeneously mixed, heterogeneously mixed, and seed-and-blanket thorium-uranium fuel cycles that rely on "in situ" use of the bred-in U-233. However, due to the higher initial enrichment required to achieve acceptable burnups, these fuels are encountering economic constraints. Thorium can nevertheless play a large role in the nuclear fuel cycle; particularly in the reduction of plutonium. While uranium-based mixed-oxide (MOX) fuel will decrease the amount of plutonium, the reduction is limited due to the breeding of more plutonium (and higher actinides) from the U-238. Here we present calculational results and a comparison of the potential burnup of a thorium-based and uranium-based mixed oxide fuel in a light water reactor (LWR). Although the uranium-based fuels outperformed the thorium-based fuels in achievable burnup, a depletion comparison of the initially charged plutonium (both reactor and weapons grade) showed that the thorium-based fuels outperformed the uranium-based fuels by more that a factor of 2; where more than 70% of the total plutonium in the thorium-based fuel is consumed during the cycle. This is significant considering that the achievable burnup of the thorium-based fuels were 1.4 to 4.6 times less than the uranium-based fuels. Furthermore, use of a thorium-based fuel could also be used as a strategy for reducing the amount of long-lived nuclides (including the minor actinides), and thus the radiotoxicity in spent nuclear fuel. Although the breeding of U-233 is a concern, the presence of U-232 and its daughter products can aid in making this fuel self-protecting, and/or enough U-238 can be added to denature the fissile uranium. From these calculations, it appears that thorium-based fuel for plutonium incineration is superior as compared to uranium-based fuel, and should be considered as an alternative to traditional MOX in both current and future reactor designs.*

I. INTRODUCTION

The use of thorium in light water reactors (LWR), and future reactor designs, presents opportunities for developing a fuel that is very resistant to nuclear weapons-material proliferation, results in a more stable and non-leachable waste form, and generates less high level waste per MW-hr generated. Because thorium-based fuel can operate cooler, and retain within the fuel more of the fission products, especially the gasses, thorium-based fuel can possibly be operated to higher burnups than uranium-based fuel.

The advantages mentioned above motivated the current work under a Laboratory Directed Research and Development (LDRD) project between the Idaho National Engineering and Environmental Laboratory (INEEL) and the Massachusetts Institute of Technology (MIT), and a subsequent separate Department of Energy (DOE) Nuclear Energy Research Initiative (NERI) award, focusing on the use of thorium-based fuel in otherwise-conventional, retrofittable, PWR fuel assemblies. Thorium fuel cycles have been studied in the past, most notably in whole-core demonstrations in Indian Point I, Elk River, and at Shippingport.[1] These cycles were directed toward the production, reprocessing, and reuse of U-233 through reactors having mainly seed-and-blanket configurations, utilizing highly enriched U-235 for startup, and achieving burnups of ~30 MWd/kg. However, circumstances have changed since then: once-through fueling is assumed; a

<20wt% U-235 anti-proliferation limitation has been imposed; and a discharge burnup approaching 60 MWd/kg has been achieved in uranium-fueled LWRs, with further increases in prospect. More recent studies[2,3] of thorium fuel cycles have been directed toward homogeneously mixed, heterogeneously mixed, and seed-and-blanket thorium-uranium fuel that rely on "in situ" use of the bred-in U-233. However, due to the higher initial enrichment required to achieve acceptable burnups, these fuels are proving less economical than UO_2 fuel.

Nevertheless, thorium may play a large role in the nuclear fuel cycle; particularly in the reduction of plutonium. In addition to the stockpiles of weapons grade plutonium, it has been estimated that there are approximately 1500 tonnes of plutonium in spent fuel from LWRs. While uranium-based mixed-oxide (MOX) fuel will decrease the amount of plutonium, the reduction is limited due to the breeding of more plutonium (and higher actinides) from the U-238. The uranium fuel cycle that is currently used contains mostly U-238, which will go through the following reaction upon capture of a neutron:

$$^{238}U(n,\gamma) \xrightarrow[\substack{\sigma_\gamma=2.68b,\\ I_{resonance}=277b}]{} {}^{239}U$$

$$^{239}U \xrightarrow[\beta^-(t_{1/2}=23.47min)]{} {}^{239}Np$$

$$^{239}Np \xrightarrow[\beta^-(t_{1/2}=2.355days)]{} {}^{239}Pu$$

$$^{239}Pu \xrightarrow[\substack{\sigma_f=750b\\ \sigma_\gamma=271b}]{} fission$$

Subsequent neutron captures will produce the higher isotopes of plutonium, which eventually result in the production of the higher actinides (e.g., Am, Cm, etc.). Note that Np-237, also a long-lived actinide, is a daughter nuclide in the Pu-239 chain; a result of the decay of Am-241.

On the other hand, the thorium chain produces the following:

$$^{232}Th(n,\gamma) \xrightarrow[\substack{\sigma_\gamma=7.37b,\\ I_{resonance}=85b}]{} {}^{233}Th$$

$$^{233}Th \xrightarrow[\beta^-(t_{1/2}=22.3min)]{} {}^{233}Pa$$

$$^{233}Pa \xrightarrow[\beta^-(t_{1/2}=27.0days)]{} {}^{233}U$$

$$^{233}U \xrightarrow[\substack{\sigma_f=531b\\ \sigma_\gamma=46b}]{} fission$$

where plutonium and the higher minor actinides are not produced. While U-233 is a proliferation concern, the presence of U-232 adds a measure of self-protection, and the U-233 can also be managed by denaturing, such that the U-233 to U_{total} ratio is <12% by adding a small amount of natural uranium to the thorium.

In this paper we present the use of a thorium-based mixed oxide fuel, and its potential benefits to decreasing the current inventory of separated plutonium (both reactor and weapons grade). In addition, use of a thorium-based fuel could also be used as a strategy for reducing the amount of long-lived nuclides, and thus the radiotoxicity, in spent nuclear fuel.

II. OBJECTIVE

The objective of this work is to verify that a thorium-based fuel could be used to efficiently burn plutonium, and to compare this capability with that of currently proposed MOX fuel (uranium-based). It is thought that the use of thorium in a MOX fuel will:

- reduce the plutonium content at a faster rate than traditional uranium MOX fuel,
- reduce the production of higher actinides during the cycle, thus reducing the repository requirements,
- improve the thermal conductivity of the fuel,
- retain fission gases better,
- and be a more stable waste form for direct disposal in the repository.[a]

In this work, we concentrated on the first two items. Future work will address the other items, and include the use of minor actinides in the fuel as a way to decrease other long-lived isotopes and further ease repository requirements.

III. ANALYSIS TOOLS AND FUEL PARAMETERS

The current work uses the MOCUP[4] code to analyze the reactivity characteristics and isotopic concentrations of unit fuel pins/cells, with 17 actinides and 41 fission products being tracked through the MCNP[5] portion of the analysis. MCNP is a well-known Monte Carlo code capable of calculating fluxes, reaction rates, and eigenvalues in general, 3-D geometry using continuous cross-section data. ORIGEN[6] uses a matrix exponential method to calculate the generation and depletion of isotopes, or elements, in a given neutron flux. MOCUP takes specific output data (including cross-section data, fluxes, and reaction rates) from MCNP and passes it to ORIGEN, where new isotopic information is generated and passed back to MCNP for the next calculation. This gives

[a] Thorium dioxide (ThO_2) is the highest oxide of thorium, while uranium dioxide (UO_2) will further oxidize to U_3O_8.

time dependent information about the reactivity swing and isotopics for the specified problem. MOCUP has been successfully benchmarked against other codes for all-uranium, MOX, and thorium-based LWR fuels.[2,3,7,8]

A 17x17 PWR lattice is assumed for the pin cell parameters, where the thorium and uranium MOX fuels are charged with 4% plutonium. The fuel parameters are summarized in Table 1.

Table 1. Fuel parameters for pin cell.

Parameters	Values (hot full power)
Fuel Temperature (K)	900
Fuel Density	94% of theoretical
Fuel Radius (cm)	0.41274
Clad Inner Radius (cm)	0.41896
Clad Outer Radius (cm)	0.47609
Pin Pitch (cm)	1.2626
Fuel Constituents	
Enrichment	4% Pu
^{239}Pu wt% (reactor grade)	58%
^{239}Pu wt% (weapons grade)	94%

A schematic cross section of the fuel pin used in the calculations can be seen in Figure 1. Note that this pin cell is 4 cm high, and reflective boundaries are used in the MCNP portion of the calculations thereby eliminating all leakage. This particular model has agreed well with infinite lattice calculations, as compared to other infinite lattice burnup codes.[7]

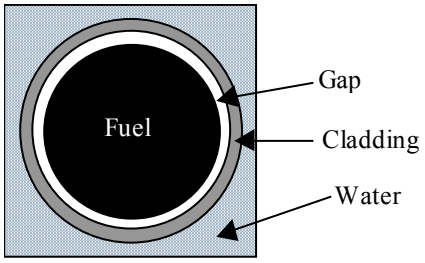

Figure 1. Cross section of pin cell (not to scale).

However, because these calculations were based on an infinite lattice, leakage will need to be taken into account when calculating the discharge burnup of the fuel. This is discussed in the next section.

IV. REACTIVITY-LIMITED BURNUP

The discharge burnup was based on a 3-batch fuel cycle, which can be calculated from the following equation:[9]

$$B_D = B_1 \cdot \frac{2n}{n+1}$$

where B_1 is the first batch burnup (chosen to be at $k_\infty \approx 1.03$ to account for leakage), n is the number of batches (3 in this case), and B_D is the discharge burnup. Figure 2 compares the reactivity curves of the different fuels, where the x-axis is the burnup in energy per unit mass, and the y-axis is the infinite neutron multiplication factor, or reactivity.

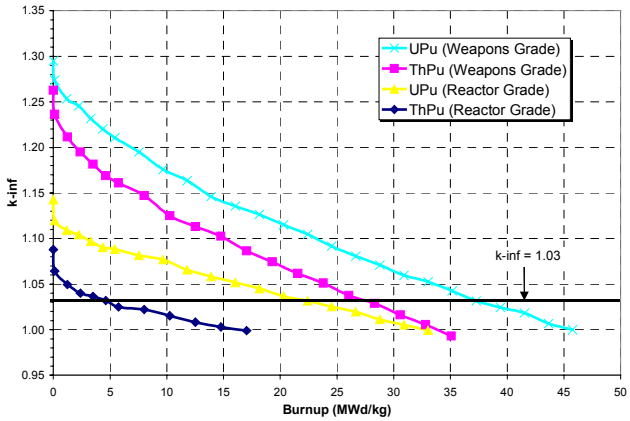

Figure 2. Reactivity-limited burnup comparison.

The reactor grade thorium-based fuel has the smallest discharge burnup at ~8 MWd/kg, while the weapons grade uranium-based fuel has the largest burnup at ~57 MWd/kg. While the reactor grade thorium-based fuel has a very low burnup, note that this study only encompassed a 4% plutonium loading in all cases, regardless of the matrix or plutonium grade. The B_1 for the uranium-based fuel with reactor grade plutonium is more than 4 times larger than the thorium-based fuel, and about 35% larger for the weapons grade plutonium (also in favor of uranium). Thus, for reactor grade plutonium, use of a uranium-based fuel will give more than 4 times the burnup of the thorium-based fuel, while use of weapons grade plutonium increases the burnup by 35% over a similar thorium-based fuel. This can be remedied by increasing the plutonium content in the thorium-based fuel. However, even at lower burnup, the plutonium vector and total plutonium that remains at discharge favors the thorium-based fuel. This will be discussed in a later section of this paper.

V. SPECTRAL DIFFERENCES

In previous studies of thorium-uranium fuel,[3,10] it was shown that spectral and shielding effects dominated the achievable burnup of the fuel. The difference between the former studies and those presented here is the fissile material loading; where this study uses reactor or weapons grade plutonium rather than U-235 enrichment. Because of the use of plutonium, one would expect a difference in the neutron spectrum regardless of the main fuel matrix (thorium or uranium).

Figures 3-6 compare the beginning-of-life (BOL) and end-of-life (EOL) spectrum of both the thorium and uranium-based fuels with respect to a 4.5% enriched (U-235) UO_2 fuel, where the x-axis is the neutron energy (in MeV) and the y-axis is the normalized flux based on 504 lethargy steps. Of particular interest in these figures is the highly depressed thermal flux in all of the cases at BOL. This is due to the large capture and fission cross sections of the plutonium isotopes, where the Pu-239 capture and fission cross sections are 2.7 and 1.3 times higher than the U-235 cross sections, respectively. In addition, the other isotopes of plutonium also have high capture and/or fission cross sections (e.g., Pu-241 fission cross section is 1009 barns).

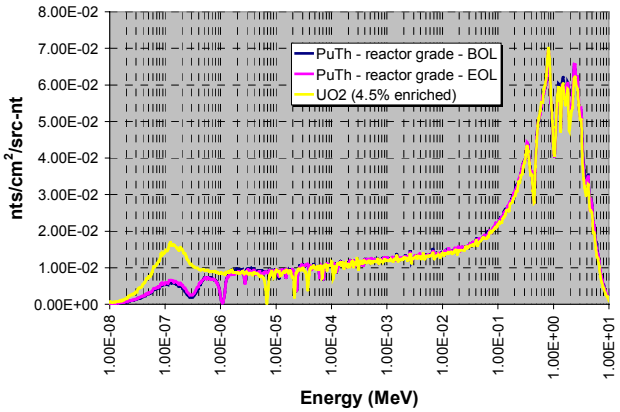

Figure 3. Neutron spectrum of thorium fuel (reactor grade plutonium) and U-235 enriched UO_2.

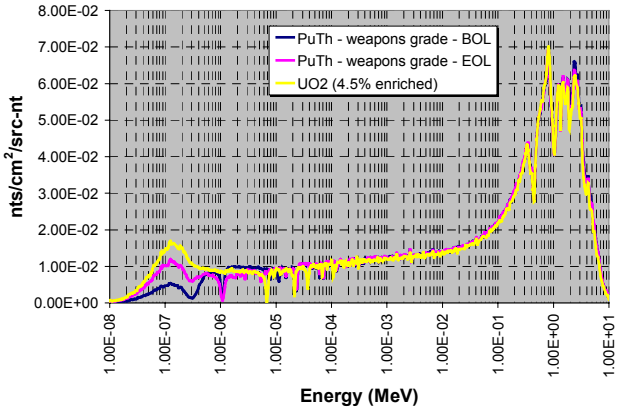

Figure 4. Neutron spectrum of thorium fuel (weapons grade plutonium) and U-235 enriched UO_2.

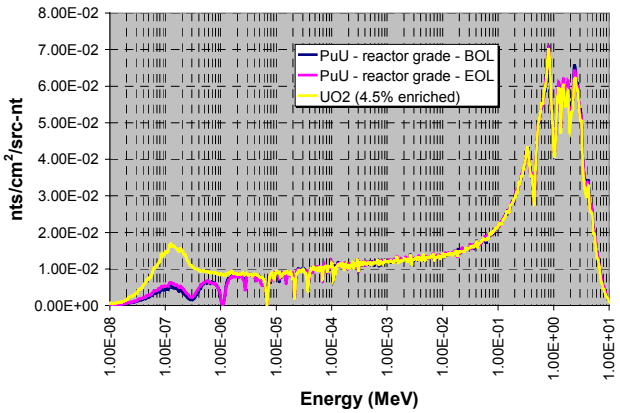

Figure 5. Neutron spectrum of uranium fuel (reactor grade plutonium) and U-235 enriched UO_2.

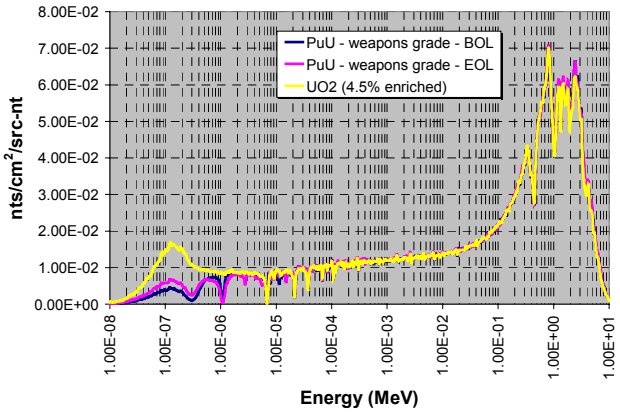

Figure 6. Neutron spectrum of uranium fuel (weapons grade plutonium) and U-235 enriched UO_2.

One can see from Figure 4 (thorium-based fuel using weapons grade plutonium) that at the EOL, the thermal flux has increased by a factor of 2. This indicates that the thermal absorbers, particularly plutonium, have been depleted significantly. When comparing this spectrum with the uranium-based fuels (see Figures 5-6), one can see only a slight increase in the thermal flux at EOL, indicating a slower decrease in the absorbers than with the thorium-based fuel, or a smaller reduction in the plutonium.

Unfortunately, the thorium-based fuel using reactor grade plutonium (see Figure 3) does not show the same increase in thermal flux at the EOL. This is due primarily to the relatively small (20%) reduction in plutonium at the EOL, and very low discharge burnup of this fuel (~8 MWd/kg).

VI. PLUTONIUM ISOTOPIC COMPARISON

As was stated earlier, the use of a thorium-based MOX fuel appears to have an advantage in plutonium destruction as compared to the uranium-based MOX.

Figures 7-8 show the plutonium isotopic concentrations (in g/cm^3 in the fuel) at several burnups. These burnups were chosen based on the calculated discharge burnup, where there are two entries for the fuel containing weapons grade plutonium. One is at the same discharge burnup as the reactor grade plutonium cases, and the other is the discharge burnup of the fuel containing weapons grade plutonium. This was done to compare the difference in isotopic concentrations at the same burnup, given that the reactor grade plutonium cases have smaller burnups.

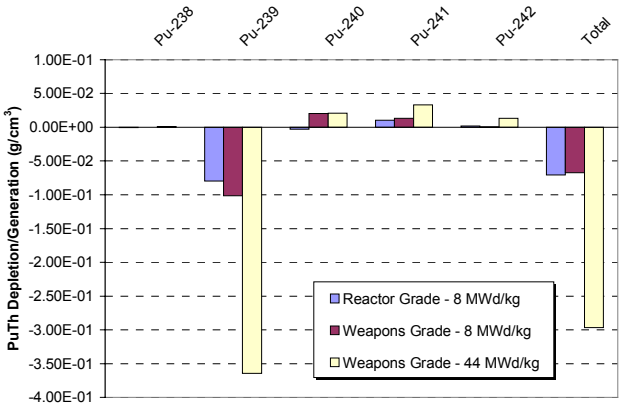

Figure 7. Average plutonium depletion/generation for thorium-based fuels.

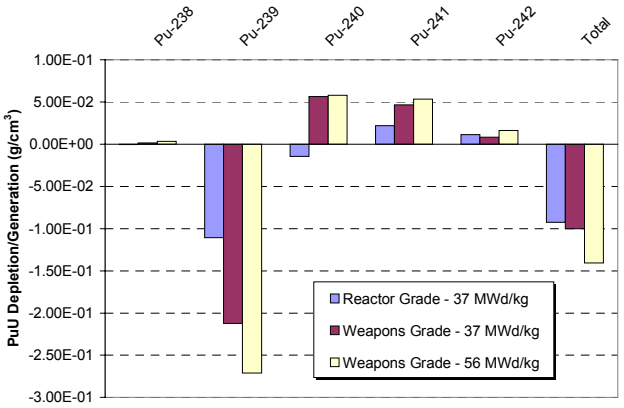

Figure 8. Average plutonium depletion/generation for uranium-based fuels.

In the thorium-based cases (see Figure 7), the difference in isotopic concentrations at 8 MWd/kg burnup are small, while the difference between the 8 and 44 MWd/kg burnups are very large. If the thorium-based fuel containing the reactor grade plutonium could be driven to the same burnup as the weapons grade case, one would expect the same total plutonium reduction. However, the plutonium isotopic fractions are different due to the different BOL fractions, i.e., the weapons grade plutonium cases are approximately 94% Pu-239, while the reactor grade is approximately 60% Pu-239 at BOL. Although the fission cross section for Pu-239 is high, so is the capture cross section. Thus, a higher concentration of Pu-239 will also produce elevated concentrations of the higher plutonium isotopes. The isotopic fractions at BOL and EOL will be shown later in this section.

Figure 9 shows the comparison of thorium and uranium-based fuels, where only the isotopic fraction concentrations at discharge burnup (i.e., EOL) are given.

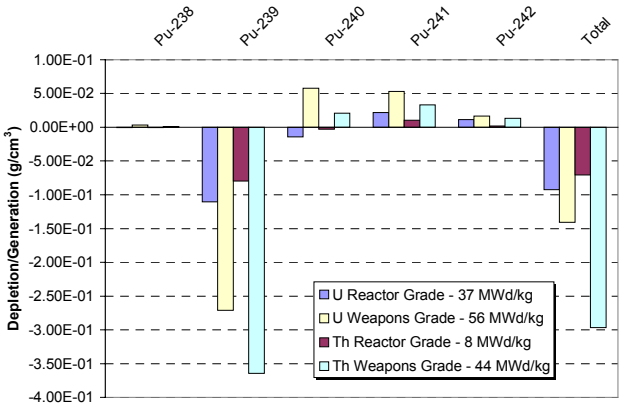

Figure 9. Average plutonium depletion/generation.

Of particular interest are the Pu-239 concentrations and the total plutonium concentration. Although the uranium-based fuel containing reactor grade plutonium can achieve a burnup that is ~4.6 times greater than the thorium-based fuel, the Pu-239 and total plutonium destruction in the uranium-based fuel is only 30% better. In the weapons grade plutonium cases, the uranium-based fuel has a burnup that is ~1.3 times greater than the thorium-based fuel, but destroys 30% less Pu-239 and less than half the total plutonium of the thorium-based fuel. Also note the larger increase in Pu-240 and Pu-241 in the uranium-based fuel as compared to the thorium-based fuel. While there is a total net destruction of plutonium in the uranium-based fuel, the destruction rate (per MWd/kg) is much slower than in the thorium-based fuel due to the plutonium bred in the uranium-based fuel.

Table 2 shows the plutonium isotopic fractions as a percent of the total plutonium present in the fuel at the discharge burnup.

Table 2. Plutonium isotopic fractions and total plutonium.

Fuel and Pu Grade	Burnup (MWd/kg)	Isotopic Fraction					Total Pu (g/cm^3)
		Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	
Th -reactor	0	2.00%	58.00%	26.00%	10.00%	4.00%	0.422
Th -reactor	8	2.18%	47.03%	30.45%	15.02%	5.31%	0.351
Th - weapon	0	0.01%	93.80%	5.81%	0.35%	0.02%	0.422
Th - weapon	44	0.83%	24.84%	36.14%	27.64%	10.56%	0.125
U -reactor	0	2.00%	58.00%	26.00%	10.00%	4.00%	0.447
U -reactor	37	2.37%	41.92%	28.73%	18.73%	8.26%	0.354
U - weapon	0	0.01%	93.80%	5.81%	0.36%	0.02%	0.447
U - weapon	56	1.07%	48.34%	27.35%	17.90%	5.35%	0.306

As compared to previous work,[2,3] the Pu-238 fractions are quite small. However, there is a marked reduction in the Pu-239 fractions in all cases, where the weapons grade plutonium cases have the most dramatic drops. However, note again that the total plutonium (and thus the Pu-239) reduction highly favors the thorium-based fuel.

VII. CONCLUSIONS

In an effort to further enhance the plutonium destruction capability of MOX fuel, the reactivity and plutonium isotopic concentrations of thorium-based MOX fuel were studied and compared to uranium-based fuel. Thorium has many attractive properties as a fuel, including better thermal conductivity than uranium, better stability as a waste form compared to uranium, and will produce less higher actinides than uranium fuel. Both reactor and weapons grade plutonium were used, at 4wt% of the heavy metal. Because a weight percent was used, there was more plutonium in the uranium-based fuels due to the higher density of uranium (as compared to thorium). Future calculations will adjust for the density difference by adding more plutonium in the thorium-based fuels.

When comparing the reactivity-limited burnup of each fuel, the uranium-based fuels outperformed the thorium-based fuels by 1.3 to 4.6 times. However, the plutonium destruction rate per MWd-cm^3 is much higher in the thorium-based fuels; 3.5 times better in the reactor grade plutonium case, and 2.7 times better in the weapons grade plutonium case (as can be seen in Figures 7-9, and Table 2). In the case of U-233 production in the thorium-based fuels, quantitative results were not obtained. However, due to the lower thermal neutron flux as compared to typical UO_2 (and even homogeneous ThO_2-UO_2 fuel[3,10]), the conversion of thorium should be minimized. Nonetheless, further analysis of the uranium isotopic fractions (particularly the U-232 content) and total amounts produced is needed, although it is expected that the uranium will be >99% U-233 in the fuel used for the studies performed and presented here.

From these calculations, it appears that thorium-based fuel for plutonium incineration is superior as compared to uranium-based fuel, and should be considered as an alternative to traditional MOX in both current and future reactor designs. Future work on thorium-based MOX fuels will include: quantifying the heating and power profiles to verify that safety parameters can be met; fuel performance modeling for both steady-state and transient conditions; manufacturing and fabrication feasibility studies; economic analysis and comparison for the fuel and fuel cycle; and eventual irradiation and post-irradiation-examination (PIE) of test pins and lead assemblies if the previous studies show promise.

ACKNOWLEDGMENTS

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NOMENCLATURE

b	Barns (cm^2)
B_D	Discharge burnup
BOL	Beginning-of-life
B_1	First batch burnup
DOE	Department of Energy
EOL	End-of-life
INEEL	Idaho National Engineering and Environmental Laboratory
$I_{resonance}$	Integral resonance cross section (barns)
LWR	Light water reactor
MCNP	Monte Carlo N-Particle computer code
MeV	Million electron-volts
MIT	Massachusetts Institute of Technology
MOCUP	MCNP-ORIGEN Coupled Utility Program
MOX	Mixed-oxide
MWd-cm^3	Megawatt-days per cubic centimeter
MWd/kg	Megawatt-days per kilogram
MW-hr	Megawatt-hours
n	Integer
NERI	Nuclear Energy Research Initiative
ORIGEN	Oak Ridge Isotope Generation and Depletion Code
PIE	Post irradiation examination
PWR	Pressurized water reactor
U_{total}	Total of all uranium isotopes
wt%	Weight percent
σ_f	Thermal fission cross section (barns)
σ_γ	Thermal capture cross section (barns)

REFERENCES

1. X. ZHAO, M. J. Driscoll, and M. S. Kazimi, "Rationale for Reconsidering the Thorium Cycle in Light Water Reactors", *Transactions of the American Nuclear Society*, Vol. **80**, pp. 43-44, American Nuclear Society, Boston, MA, USA, June (1999).

2. P. E. MACDONALD et al., "Advanced Proliferation Resistant, Lower Cost, Uranium-Thorium Dioxide Fuels for Light Water Reactors," Idaho National Engineering and Environmental Laboratory, INEEL/EXT-2000-01217, August (2000).

3. P. E. MACDONALD et al., "Advanced Proliferation Resistant, Lower Cost, Uranium-Thorium Dioxide Fuels for Light Water Reactors," Idaho National Engineering and Environmental Laboratory, INEEL/EXT-01-00804, August (2001).

4. R. L. MOORE, B. G. Schnitzler, C. A. Wemple, R. S. Babcock, and D. E. Wessol, "MOCUP:MCNP-ORIGEN2 Coupled Utility Program," Idaho National Engineering Laboratory, INEL-95/05231995, September (1995).

5. J. F. BRIESMEISTER, Ed., "MCNP™ - A General Monte Carlo N-Particle Transport Code (version 4b)," Los Alamos National Laboratory, LA-12625-M, March (1997).

6. A. G. CROFF, Ed., "A User's Manual for the ORIGEN2 Computer Code (version 2.1)," Oak Ridge National Laboratory, ORNL/TM-7175, July (1980).

7. X. ZHAO, E. E. Pilat, K. D. Weaver, and P. Hejzlar, "A PWR Thorium Pin Cell Burnup Benchmark," *Proceedings of PHYSOR 2000*, American Nuclear Society, Pittsburgh, PA, USA, May (2000)

8. X. ZHAO, K. D. Weaver, Y. Xu, and T. J. Downar, "Comparison of Computer Code Predictions for ThO_2 Pincell Burnup," *Transactions of the American Nuclear Society*, Vol. **83**, pp. 197-199, American Nuclear Society, Washington DC, USA, November (2000).

9. M. J. DRISCOLL, T. J. Downar, and E. E. Pilat, *The Linear Reactivity Model for Nuclear Fuel Management*, American Nuclear Society, LaGrange Park, IL, USA (1990).

10. K. D. WEAVER and J. S. Herring, "The Effect of the Hydrogen to Heavy Metal Ratio (H/HM) on Reactivity and Discharge Isotopics of Homogeneous Thoria-Urania Fuel," *Proceedings of the 10th International Conference On Nuclear Engineering (ICONE-10)*, CD-ROM, American Society of Mechanical Engineers, Arlington, VA, USA, April (2002).